Testing JEFF-3.1.1 and ENDF/B-VII.1 Decay and Fission Yield Nuclear Data Libraries with Fission Pulse Neutron Emission and Decay Heat Experiments

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The aim of this work is to test the present status of Evaluated Nuclear Decay and Fission Yield Data Libraries to predict decay heat and delayed neutron emission rate, average neutron energy and neutron delayed spectra after a neutron fission pulse. Calculations are performed with JEFF-3.1.1 and ENDF/B-VII.1, and these are compared with experimental values. An uncertainty propagation assessment of the current nuclear data uncertainties is performed.

I. INTRODUCTION

To perform calculations of decay heat and delayed neutron emission an updated version of ACAB code [1] is used. ACAB code is a computer program designed to perform activation and transmutation calculations for nuclear applications. It includes a sensitivity analysis tool to identify critical radionuclides and pathways contributing to their production, and its uncertainty capability is used to assess the impact of nuclear data uncertainties on activation-related quantities. For decay heat, beta, gamma and total contributions are computed to compare ENDF/B-VII.1 [2] and JEFF-3.1.1 [3] decay and fission yield data libraries. An updated JEFF-3.1.1 decay data library is presented in this work including the recent TAGS experimental data [4]. However, ENDF/B-VII.1 already includes these values. For delayed neutron emission, each isotope able to emit delayed neutrons following beta decay is taken into account. These results are compared with the ones furnished by libraries themselves through the adjusted *Keepin formulation*, which is an experimental-based approach with reliable results used since 50's in nuclear reactor design.

To perform an uncertainty analysis, different error propagation techniques (adjoint/forward sensitivity analysis procedures and Monte Carlo technique) can be used. In this work, a Monte Carlo technique [5] is used defining a joint probability distribution of the nuclear data uncertainties. A log normal probability density function is assumed, $log(\alpha/\hat{\alpha}) \rightarrow N(0,V)$, where V is the variance matrix of the nuclear data relative error, α=($T_{1/2}$, branching ratio, Energy released by decay - Qrec, fission yield) is a random vector of nuclear data involved in the problem, and $\hat{\alpha}$, is the best-estimated nuclear data vector. This Monte Carlo techniques enables one to investigate the importance of nuclear data uncertainties in decay and fission yields data libraries on decay heat and delayed neutron emission.

II. FISSION PULSE DECAY HEAT

For rapid reactor transients, the prediction of decay heat is important in the range of seconds to minutes. In this work, we assess the current status of Evaluated Nuclear Decay and Fission Yield Data Libraries to predict the decay heat rate and beta-gamma contributions. The decay heat as a function of time is given by

$$DH(t) \; = \; \sum_i \lambda_i N_i(t) Q_i, \qquad (1)$$

where, λ_i is the decay constant of isotope-i, $N_i(t)$ is the number of isotope-i present at time-t after irradiation, and Q_i is the energy released per decay of a nuclide-i.

Fig. 1 shows the total (electromagnetic and light particles) decay heat for thermal neutron induced fission on ^{239}Pu following a fission burst. Good agreement of ENDF/B-VII.1 with the compilation of experimental data provided by Tobias [6] is found. A significant improvement in the total decay heat prediction is obtained if the total absorption gamma spectroscopy (TAGS) [4] measurements are included in JEFF-3.1.1, providing a better description of the electromagnetic decay heat component. It should be remarked that Tobias fit is conservative relative to many experiments conducted in the past. In addition, a comparison of the calculated total decay heat with other measurements [7–10] is illustrated, show-

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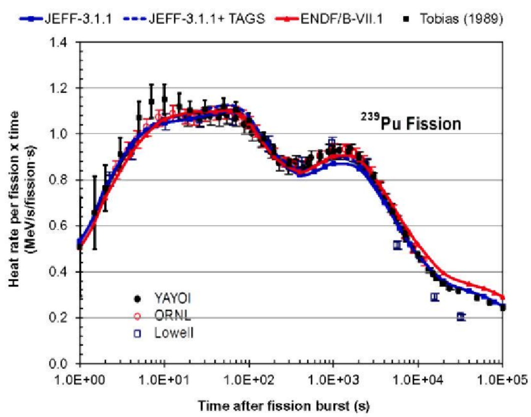

FIG. 1. Experimental measurements of ^{239}Pu fission decay heat for a fission pulse with thermal neutrons: ORNL [7], Lowell [10], and YAYOI [8, 9]. Results with JEFF-3.1.1 (+ TAGS) and ENDF/B-VII.1 are predicted with ACAB code.

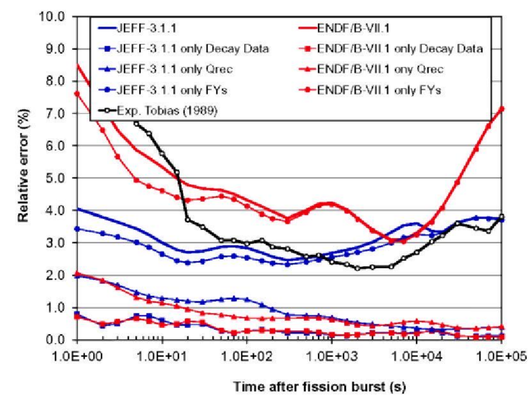

FIG. 2. Predicted relative errors (in %) due to all nuclear data uncertainties propagated together and individually, compared to experimental uncertainty [6] of total decay heat (β+γ) for thermal neutron fission burst of ^{239}Pu.

ing a better agreement of calculations at short times (1-10 seconds).

It should be noted that JEFF-3.1.1 has many isotopes with unknown decay energy uncertainties for beta and gamma emission. These isotopes represent about ~17% of total decay heat for times shorter than 2000 s, for these isotopes a value of decay energy uncertainty of 15% is assumed. In Table I, a list of the most important contributors at 1000 s after a fission burst is shown; isotopes measured by Ref. [4] are referenced in the Table. To address a sensitivity analysis, a preliminary pathway analysis is performed. For instance, the most important pathways to generate ^{104}Tc are: ^{239}Pu $\longrightarrow$ ^{104}Mo $\longrightarrow$ ^{104}Tc (69%) and ^{239}Pu $\longrightarrow$ ^{104m}Nb $\rightarrow$ ^{104}Mo $\rightarrow$ ^{104}Tc (10%), ^{239}Pu $\longrightarrow$ ^{104}Nb $\rightarrow$ ^{104}Mo $\rightarrow$ ^{104}Tc (10%), ^{239}Pu $\rightarrow$ ^{104}Tc (10%). Consequently, a sensitivity analysis should take into account a large set of different decay and fission yield nuclear data contributions. This problem is overcome using a Monte Carlo technique.

In Fig. 2, uncertainty in total decay heat due to the current uncertainties in these Nuclear Data Libraries is predicted. To perform this uncertainty calculation a Monte Carlo technique is used with a total random sampling of 1000 histories of different Decay/Energy and Fission Yield Data Libraries. It can be seen that the uncertainty of fission yields is the most important contributor to the total decay heat uncertainty. The total average uncertainty prediction is ~3% with JEFF-3.1.1 and ~5% with ENDF/B-VII.1.

III. FISSION PULSE DELAYED NEUTRONS

In this section, the present status of Evaluated Decay and Fission Yield Data Libraries is assessed to predict the total delayed neutron emission, neutron emission rate and the corresponding average neutron energy. Large differences of the number of βn-emitters in these libraries are found: JEFF-3.1.1 with 241 βn-emitters, 18 β2n-emitters and only 4 β3n-emitters, and ENDF/B-VII.1 with 390 βn-emitters, 111 β2n-emitters, 14 β3n-emitters and 2 β4n-emitters. JEFF-3.1.1 is based mainly on NUBASE-2003 [11], where nuclides for which some experimental information is known are considered. ENDF/B-VII.1 supplements with recent theoretical calculations of the continuous spectrum from beta-delayed neutron emitters. To compare with the experimental values, the adjusted *Keepin formula* is used taking data from the neutron induced ENDF libraries (section MT455).

TABLE I. Total decay heat and relative contribution in % of the most important contributors to the total decay heat at 1000s after thermal neutron fission burst of ^{239}Pu.

JEFF-3.1.1 Total: 0.87 MeV/fission	%	ENDF/B-VII.1 Total: 0.91 MeV/fission	%
^{104}Tc [4]	8.6	^{104}Tc[4]	9.9
^{95}Y	5.3	^{105}Tc[4]	5.7
^{102}Tc[4]	5.2	^{95}Y	5.1
^{101}Mo	5.1	^{102}Tc [4]	5.1
^{139}Cs	4.9	^{101}Mo	4.7
^{105}Tc[4]	4.6	^{139}Cs	4.4

A. Total Delayed Neutron Emission

The total delayed neutron emission per fission can be formulated as

$$\bar{\nu}_d = \sum_i P_{ni} c_i, \qquad (2)$$

where c_i is the cumulative fission yield of isotope-i and

TABLE II. Average delayed neutron yields following a single thermal fission.

Nuclide	JEFF-3.1.1[3]	This work (JEFF-3.1.1)	This work (ENDF/B-VII.1)
^{233}U	0.72±0.01	0.72±0.06	0.74±0.06
^{235}U	1.47±0.02	1.47±0.08	1.91±0.10
^{237}Np	1.12±0.03	1.13±0.08	1.66±0.15
^{238}Pu	0.31±0.01	0.32±0.04	-
^{239}Pu	0.60±0.02	0.60±0.04	0.72±0.02
^{241}Pu	1.23±0.03	1.22±0.06	1.34±0.07
^{241}Am	0.37±0.01	0.38±0.05	0.56±0.07
^{242m}Am	0.58±0.02	0.58±0.11	0.77±0.09
^{243}Am	0.84±0.02	0.84±0.11	-
^{243}Cm	0.22±0.01	0.22±0.04	0.47±0.06
^{244}Cm	0.32±0.01	0.33±0.05	-
^{245}Cm	0.52±0.01	0.52±0.07	0.71±0.08

P_{ni} is the probability of a nuclide-i emitting a neutron as a result of a beta decay. Table II shows the average delayed neutron yields following a single thermal fission for several fissile nuclides. A large difference between JEFF-3.1.1 and ENDF/B-VII.1 is found for some isotopes. For instance, ENDF/B-VII.1 overpredicts ~23% the JEFF-3.1.1 value for ^{235}U.

The uncertainty in the calculated value $\bar{\nu}_d$ can be estimated assuming c_i and P_{ni} are independent as

$$var(\bar{\nu}_d) \;=\; \sum_i \left\{ P_{n}^2{}_i var(c_i) + var(P_{ni}) c_i^2 \right\}. \qquad (3)$$

Using Eq. 3, relative errors for these fissile nuclides are predicted in Table II. It can be noticed a large discrepancy between Ref. [3] (1st column) and this work using JEFF-3.1.1 (2nd column). In this work, JEFF-3.1.1 and ENDF/B-VII.1 show a relative error of ~5%.

For ^{235}U thermal fission, the most important contributors are: ^{137}I (15.8%), ^{89}Br (13.0%), ^{94}Rb (10.3%), ^{88}Br (8.3%), ^{90}Br(8.2%), ^{138}I(5.3%), ^{98m}Y(4.6%), ^{139}I(4.0%) and ^{95}Rb(3.9%).

B. Delayed Neutron Emission Rate

The delayed neutron emission per unit time after a pulse fission from the activity of precursors is given by

$$n_{emit}(t) \;=\; \sum_i P_{ni} \lambda_i N_i(t), \qquad (4)$$

where λ_i is the decay constant of isotope-i, $N_i(t)$ is the number of isotope-i present at time-t after irradiation and P_{ni} is the probability of a nuclide-i emitting a neutron as a result of a beta decay. Being, $N_i(0) = \gamma_i$, for only 1 fission, and γ_i is the independent fission yield.

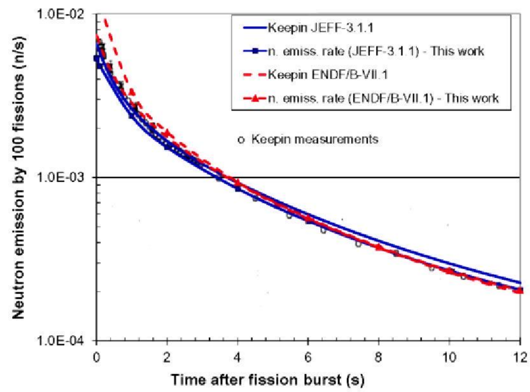

FIG. 3. Comparison of delayed neutron emission rate, n_{emit}(t), calculated using *Keepin 6/8-group formula* and the Decay and Fission Yield Data after a fission pulse in ^{235}U with thermal neutrons. Experimental values taken from Ref. [12]

According to the *Keepin formula*, the $n_{emit}(t)$ function can be approximated with a sum of 8 (in JEFF-3.1.1) or 6 (in ENDF/B-VII.1) exponential functions as

$$n_{emit}(t) \;=\; n_f \;\sum_i \nu_{di} \lambda_{di} e^{-\lambda_{di} t}, \qquad (5)$$

where ν_{di} is the yield of the i-th delayed-neutron group, λ_{di} is the decay constant of the i-th delayed-neutron group, and n_f is the number of fissions.

In Fig. 3, a comparison of the neutron emission rate for ^{235}U-thermal neutron fission using both Decay/Fission Yield Data and Cross-Section Data (MT455) is shown. Results with the *Keepin formula* using MT455 show good agreement between JEFF-3.1.1 and ENDF/B-VII.1. Calculations performed with ENDF/B-VII.1 using Eq. 4 induce a significant overestimation. By contrast, JEFF-3.1.1 predicts a slight underestimation of the results.

At shutdown, isotopes with a relative contribution greater than 5% to the total neutron emission for JEFF-3.1.1 are: ^{95}Rb (18.8%), ^{90}Br (7.9%), ^{91}Br (7.2%), ^{94}Rb (6.7%), ^{96}Rb (5.8%) and ^{89}Br (5.4%), with a total emission rate of 0.0054 n/s/100 fissions. For ENDF/B-VII.1, the most important contributors are: ^{88}As (18.7%), ^{86}Ge (14.1%), ^{100}Rb(9.1%), ^{95}Rb (7.2%), ^{137}Sb(6.8%) and ^{131}Cd(5.7%), with a total emission of 0.017 n/s/100 fissions. ^{88}As and ^{86}Ge, the two most important contributors in ENDF/B-VII.1 are not considered in JEFF-3.1.1, where these independent fission yield values are not provided. Differences in P_{ni} values between JEFF-3.1.1 and ENDF/B-VII.1 for ^{100}Rb, ^{137}Sb and ^{131}Cd are responsible for the large discrepancy between these libraries.

The delayed neutron average energy is given in Fig. 4. With *Keepin adjustment*, calculations with JEFF-3.1.1 give good agreement with the experimental values [12]. However, a large discrepancy is found with ENDF/B-

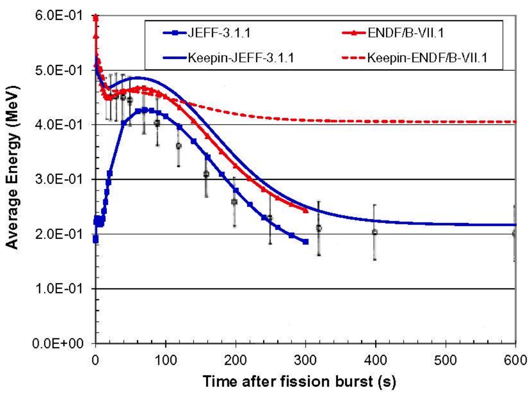

FIG. 4. Delayed neutron average energy calculated with JEFF-3.1.1 and ENDF/B-VII.1. Experimental values from Ref. [12] after a fission burst of ^{235}U with thermal neutrons.

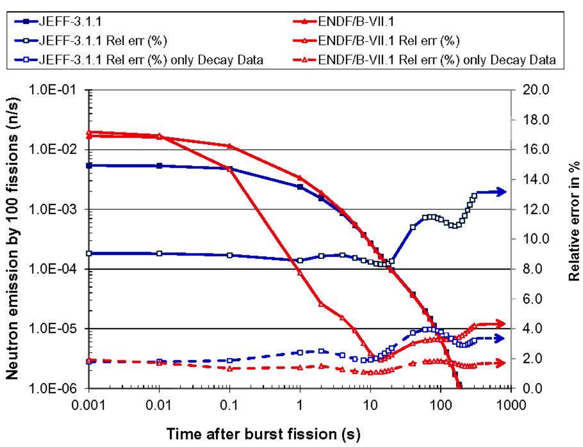

FIG. 5. Neutron Emission relative error calculated due to uncertainties in Decay Data and Independent Fission Yields after a fission pulse in ^{235}U with thermal neutrons.

VII.1. Using Eq. 4 with decay and fission yield data, more accurate results are predicted with ENDF/B-VII.1. The large discrepancy at short times for JEFF-3.1.1 is due to a lack of information on delayed neutron spectra for many important isotopes, ~35% of the total neutron contributors having no neutron spectra information.

The uncertainty analysis for neutron emission rate is presented in Fig. 5. For JEFF-3.1.1, the total uncertainty due to decay and fission yield data is 8-12% where the decay uncertainty is only 2-4%. Thus, the most important source of uncertainty is the uncertainty of fission yields. For ENDF/B-VII.1, the total uncertainty at short times is ~15%, but it decreases to ~4% for times beyond 10s after fission burst. Finally, the analysis of the neutron average energy uncertainty predicts an average uncertainty of ~6% and ~2%, for JEFF-3.1.1 and ENDF/B.VII.1, respectively.

IV. CONCLUSIONS

The present status of the Evaluated Nuclear Data Libraries JEFF-3.1.1 and ENDF/B-VII.1 is assessed in fission burst calculations for decay heat and delayed neutrons. For decay heat, the improvement of JEFF-3.1.1 with TAGS measurement produces results as good as ENDF/B-VII.1. For delayed neutron emission, large discrepancies between libraries are found for the predictions of total number of delayed neutrons. Differences in the prediction of the delayed neutron rate with decay and fission yield data against experimental adjustment with *Keepin formula* are found, showing for short times a slight underestimation with JEFF-3.1.1 and a large overestimation with ENDF/B-VII.1. Regarding average delayed neutron energy, a lack of delayed neutron spectra information in Decay Data Library JEFF-3.1.1 explains large differences at short times. Finally, it has been noticed poor results predicting the average neutron delayed energy with *Keepin formula* using ENDF/B-VII.1.

An uncertainty propagation calculation of the current nuclear data uncertainties has shown the important contribution of fission yields uncertainties both in decay heat and delayed neutron emission uncertainties.

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[1] J. Sanz *et al.*, ACAB Inventory Code for Nuclear Applications: User's Manual V.2008.
[2] M.B. Chadwick *et al.*, Nucl. Data Sheets **112**, 2887 (2011).
[3] M.A. Kellet *et al.*, JEFF Report 20, NEA/OECD, 2009.
[4] A. Algora *et al.*, Phys. Rev. Lett. **105**, 202501 (2010).
[5] O. Cabellos *et al.*, J. Korean Phys. Soc. **59**, 1268 (2011).
[6] A. Tobias, CEGB Report No. RD/B/6210/R89 (1989).
[7] J. K. Dickens *et al.*, Nucl. Sci. Eng. **78**, 126 (1981).
[8] M. Akiyama *et al.*, J. Atomic Ener. Soc. Japan **24**, 709 (1982) - in Japanese.
[9] M. Akiyama *et al.*, J. Atomic Ener. Soc. Japan **24**, 803 (1982) - in Japanese.
[10] W.A. Schier *et al.*, DOE/ER/40723-4 (1997).
[11] G. Audi *et al.*, Nucl. Phys. A **729**, 3 (2003).
[12] G. Rudstam *et al.*, J. Nucl. Energy **6**, 1 (1957).